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Personal Exposure to Volatile Organics and Other Compounds Indoors and Outdoors The TEAM (Total Exposure Assessment Methodology) Study

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16. ABSTRACT

The major objective of this study is to develop and field test a methodology for measuring individual human exposure to toxic substances. A secondary objective is to develop methods for estimating body burden with the use of biological measurements. All significant pathways of exposure are addressed. In Phase 1 of the study, sampling and analytical protocols were tested for volatile organic compounds, organochlorine pesticides, metals, and polyaromatic hydrocarbons. In Phase 2, exposure through air and drinking water and excretion rates through inhaled breath were measured for a statistically valid sample population. It was determined that personal air median concentrations ranged from 40 to 320% higher than outdoor fixed air concentrations. Correlations between personal and outdoor samples were poor. It was concluded that personal air, indoor air, or breath measurements are far superior to outdoor measurements for estimating exposure. The study is currently entering its third phase.

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Personal Exposure to Volatile Organics and Other Compounds Indoors and Outdoors -- The TEAM Study

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Introduction

For the past 3 years, the U.S. Environmental Protection Agency (EPA) has sponsored the Total Exposure Assessment Methodology (TEAM) Study (Pellizzari, 1980; Pellizzari, 1981). The overall objective of the study is to develop and field-test a methodology for measuring individual human exposure to a number of toxic substances through all important pathways (such as air, food, and drinking water). A secondary objective is to develop methods for estimating body burden by biological measurements (breath, blood, and urine). The study is presently entering its third and final phase.

Phase I: Methodology Development and Evaluation

The objective of the first phase was to test a number of sampling and analytical protocols for four groups of chemicals: volatile organic compounds (such as benzene and chloroform); organochlorine pesticides (such as DDT) and PCB's; metals (lead, cadmium, arsenic); and polyaromatic hydrocarbons (PAH's). An initial field study concentrated only on the first group of compounds -- volatile organics (Table 1) -- in air, drinking water, blood, urine, and exhaled breath (Wallace, 1982). A personal monitor (Fig. 1) employing Tenax sorbent with a Dupont pump was supplied to 11 students at Lamar University in Beaumont, Texas and 6 students at the University of North Carolina at Chapel Hill. They wore the monitor for 5-9 hours while attending classes. At the end of that period, they gave a sample of their breath, collected by a spirometer with two attached Tenax cartridges (Fig. 2). Two to three drinking water samples were also collected during the sampling period and analyzed by a purge-and-trap technique (Bellar, 1974). Among the interesting results of this preliminary field test were the following:

- Ten of the 15 target compounds were found in more than 50% of the air samples, and 7 of 15 in more than 50% of the breath samples.
- Geometric means for seven compounds ranged between 1 and 50 ug/m^3 both in air and in breath (Fig. 3).
- A significant relationship was noted between levels in air and breath for three compounds. The compounds and the parameters of the log-linear association were:

  Tetrachloroethylene: $\log B = 0.23 \pm 0.44 + (0.72 \pm 0.13) \log A$

  1,1,1-Trichloroethane: $\log B = -0.98 \pm 0.54 + (0.91 \pm 0.23) \log A$

  Vinylidene chloride: $\log B = -0.24 \pm 0.67 + (0.71 \pm 0.17) \log A$

  where B = breath concentration, A = personal air concentration, in ug/m^3.

- Only two compounds were primarily water-borne: chloroform and bromodichloromethane. Estimated daily intakes of each trihalomethane were at least 75% through water or other beverages and less than 25% through air.

A second and much more extensive pilot study was undertaken over a 6-month period on nine New Jersey subjects and three EPA employees in North Carolina (Pellizzari, 1982; Sparacino, 1982a,b; Wallace 1983a). The four groups of compounds mentioned earlier (VOC's, pesticides, metals and PAH's) were sought in four environmental media (air, water, food, and house dust) and in four biological media (breath, blood, urine, and hair) (Tables 2 and 3). A 72-hour sampling period for each subject was employed in July, September, and December, according to a set schedule (Table 4).

Sampling and Analytical Methdology

Volatile Organics. About a dozen compounds were sought in air, water, food, blood, breath, and urine. Personal air and breath samples employed the Tenax personal monitor and spirometer described previously. Food samples collected from grocery stores were analyzed using an experimental headspace analytical technique developed by the U.S. Food and Drug Administration (FDA) (Entz 1982). Blood and urine samples were analyzed using a technique developed by the University of Miami Medical School (Peoples 1979).

Metals. An air sample was collected either at the subject's home or at his work place (including commuting) over a 12-hour period using a personal monitor with a pump (2L/min) and Teflon filter. Analysis was by proton-induced x-ray emission (PIXE). Blood and urine samples were collected and analyzed by atomic absorption spectroscopy (AAS). Hair samples were collected from six of the 12 subjects and analyzed for the three target metals using AAS. Drinking water samples were collected at home and at work and analyzed by AAS. House dust samples were collected by vacuuming a 1-meter area in the living area (two attic samples were also taken) and analyzed by AAS.

Pesticides and PCB's. Indoor air samples were collected on a polyurethane foam (PUF) cartridge attached to the same monitor used to collect the metals. Analysis was by solvent extraction followed by gas chromatography with electron capture detection (GC/ECD). Drinking water samples were analyzed using Method 608 (solvent extraction followed by GC/ECD analysis) of the EPA Environmental Monitoring and Support Laboratory in Cincinnati (EMSL-CI). Blood and urine samples were analyzed using the extraction/GC/ECD technique developed at the EPA Health and Environmental Research Laboratory in Research Triangle Park (HERL-RTP).

PAH's. Amounts of six PAH's collected over a 12-hour period were measured on a portion of the same filter used to collect metals. Analysis was by a method developed by the National Institute for Occupational Safety and Health (NIOSH) employing extraction with cyclohexane, thin-layer chromatography, acetonitrile extraction, and reverse-phase high-performance liquid chromatography (HPLC). Drinking water samples were analyzed by an EMSL-CI method employing reverse phase HPLC and capillary GC. Dust samples were analyzed by the procedure used for air samples. Blood and urine samples were analyzed using extraction by hexane and gas chromatography with flame-ionization detection (GC/FID).

Table 1. CHEMICALS SOUGHT IN STUDY

	Chemical	Air and Breath[a]	Drinking Water, Blood, and Urine[a]	Blood[b] (Lamar only) (Dec. 79)	Blood[b] (Lamar only) (March 80)
1.	Chloroform	X	X	X	X
2.	Chlorobenzene	X	X	X	X
3.	Tetrachloroethylene	X	X	X	X
4.	1,2-Dichloroethane	X	X		X
5.	1,1,1-Trichloroethane	X	X	X	X
6.	1,1,2-Trichloroethane		X }		
7.	Trichloroethylene	X	X }	X	X
8.	Carbon tetrachloride		X }	X	X
9.	Bromodichloromethane	X	X }	X	X
10.	Vinylidene chloride	X		X	X
11.	1,1-Dichloroethane	X			
12.	1,2-Dichloropropane	X			
13.	Dibromochloromethane	X		X	
14.	Ethylene dibromide	X			
15.	m-dichlorobenzene	X			
16.	o-dichlorobenzene	X			X
17.	Benzene	X			
18.	Bromoform			X	X
19.	1,1,2,2-Tetrachloroethane			X	X
20.	Hexachloro-1,3-butadiene			X	X
21.	1,2-Dichloroethylene			X	

[a] Analyzed at Research Triangle Institute.

[b] Analyzed at University of Miami.

} Chemicals could not be separated by gas chromatographic column employed.

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Fig. 1. Tenax cartridge used in TEAM study. Dupont pump can be carried in vest pocket or attached to belt.

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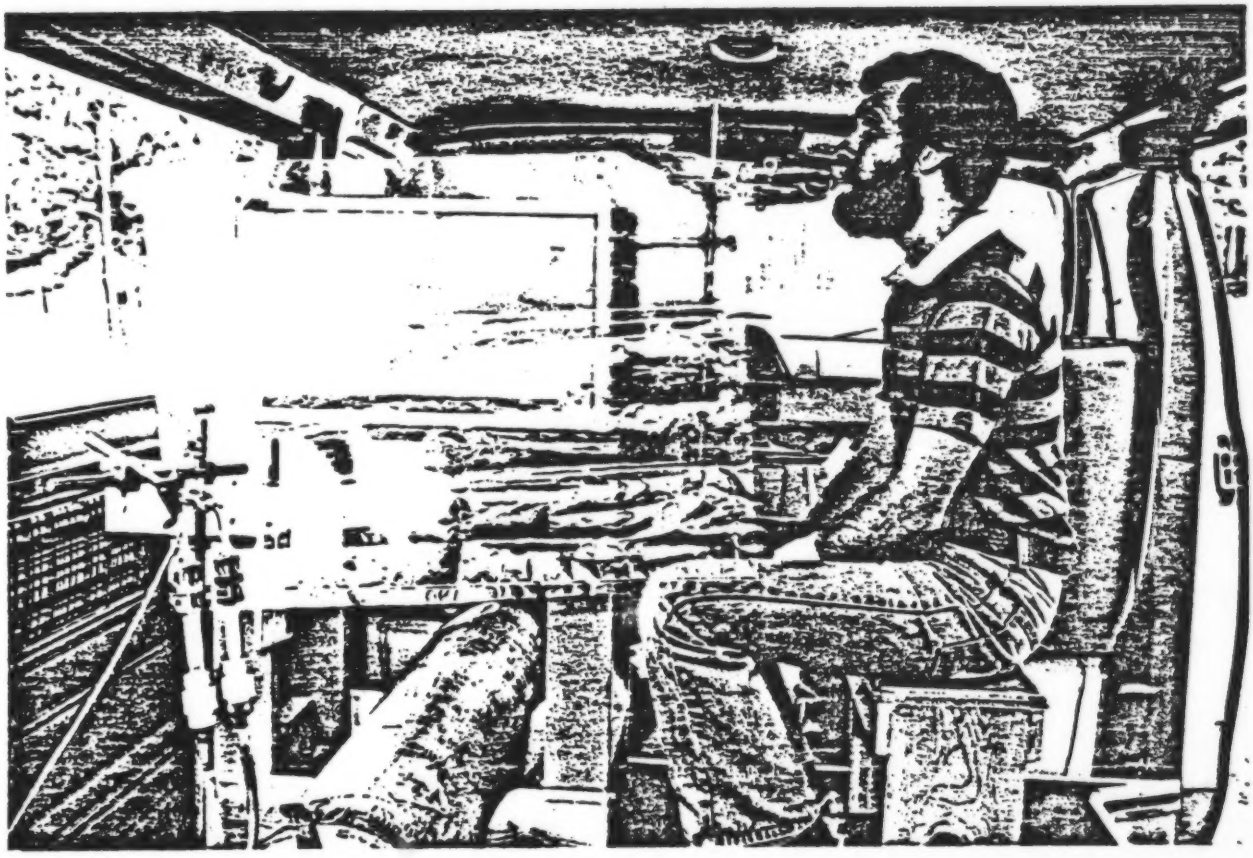

Fig. 2. Spirometer includes pure-air tank, Douglas 2-way mouthpiece, Tedlar bags, and duplicate Tenax cartridges.

Geometric Mean Concentrations of Seven Volatile Organics in Air and Exhaled Breath of Two Student Groups

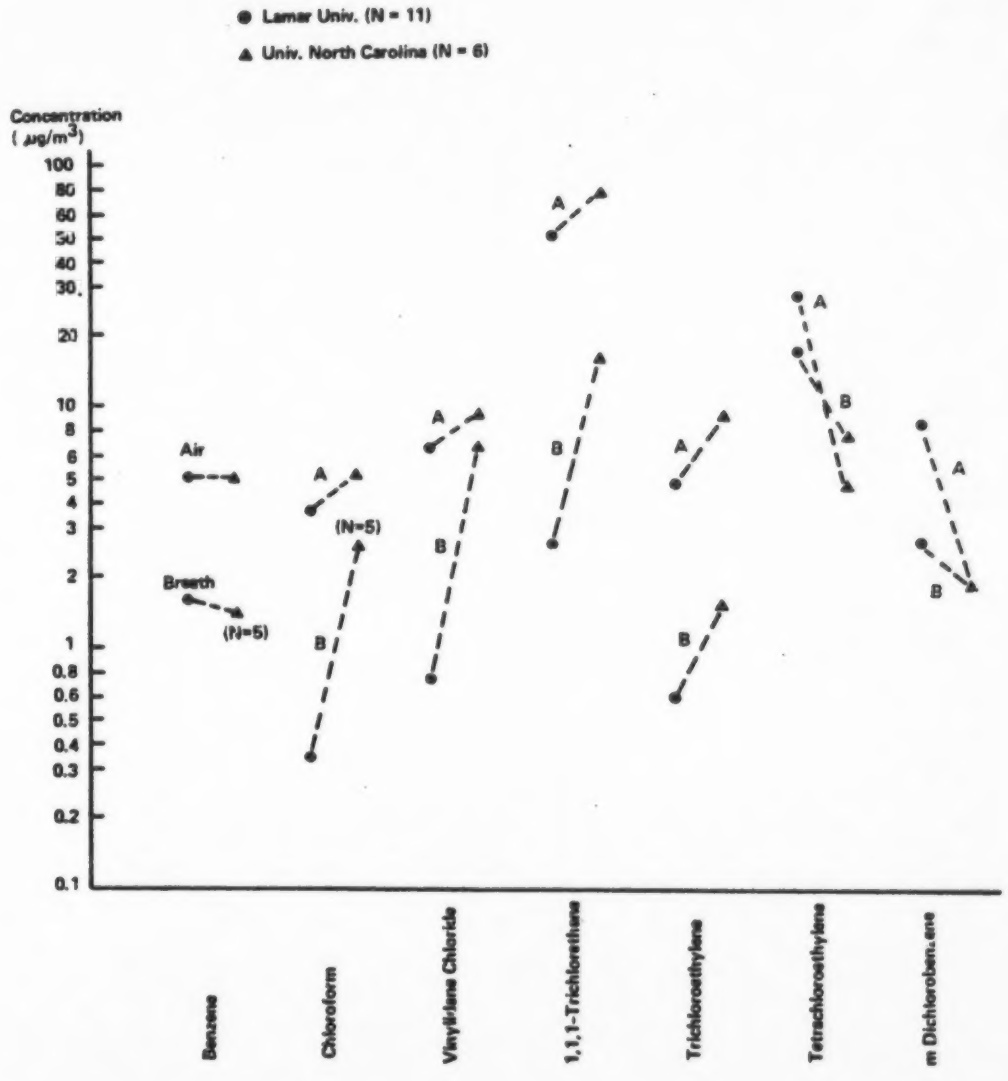

Fig. 3. Air and breath concentrations of volatile organics for students at two universities were generally similar.

A second and much more extensive pilot study was undertaken over a 6-month period on nine New Jersey subjects and three EPA employees in North Carolina (Pellizzari, 1982; Sparacino, 1982a,b; Wallace 1983a). The four groups of compounds mentioned earlier (VOC's, pesticides, metals and PAH's) were sought in four environmental media (air, water, food, and house dust) and in four biological media (breath, blood, urine, and hair) (Tables 2 and 3). A 72-hour sampling period for each subject was employed in July, September, and December, according to a set schedule (Table 4).

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Table 2. TOXIC CHEMICALS SELECTED FOR MONITORING IN ENVIRONMENTAL MEDIA (PHASE I)

Air					
Vapors	Particulate	Drinking water	Beverages	Food (tent.)	Household Dust
Benzene	Arsenic	Benzene	Chloroform	Arsenic	Arsenic
Chloroform	Cadmium	Chloroform	1,2-Dichloroethane	Cadmium	Cadmium
1,2-Dichloroethane	Lead	1,2-Dichloroethane	1,1,1-Trichloroethane	Lead	Lead
1,1,1-Trichloroethane	----------	1,1,2-Trichloroethane	1,1,2-Trichloroethane	----------	----------
Carbon tetrachloride	Benzo(a)pyrene	Carbon tetrachloride	Vinylidene chloride	α-BHC	Benzo(a)pyrene
Vinylidene chloride	Pyrene	Vinylidene chloride	Trichloroethylene	Lindane	Fluoranthene
Trichloroethylene	Chrysene	Trichloroethylene	Tetrachloroethylene	Heptachlor	Benzo(k)fluoranthene
Tetrachloroethylene	Benzo(a)anthrene	Tetrachloroethylene	Bromodichloromethane	Heptachlor epoxide	Pyrene
Bromodichloromethane	Fluoranthene	Bromodichloromethane	Chlorobenzene	Chlordane	Chrysene
Chlorobenzene	Benzo(k)fluoranthene	Chlorobenzene	Vinyl chloride	t-Nonachlor	Benzo(a)anthrene
1,1,2-Trichloroethane		Vinyl chloride		Oxychlordane	----------
----------		1,1,1-Trichloroethane		HCB	α-BHC
Vinyl chloride		----------		DDT/DDD/DDE	Lindane
----------		Arsenic		PCBs	Heptachlor
α-BHC		Cadmium			Chlordane
Lindane		Lead			HCB
Heptachlor		----------			DDT/DDD/DDE
Chlordane		Benzo(a)pyrene			PCBs
HCB		Fluoranthene			
DDT/DDD/DDE		Benzo(k)fluoranthene			
PCBs		Pyrene			
		Chrysene			
		Benzo(a)anthrene			

Table 3. TOXIC CHEMICALS SELECTED FOR MEASUREMENT IN HUMAN BODY FLUIDS AND TISSUES (PHASE I)

Breath	Blood	Urine	Mother's Milk (tent.)	Hair
Benzene	Benzene	Arsenic	Benzene	Arsenic
Chloroform	---	Cadmium	Chloroform	Cadmium
1,2-Dichloroethane	Chloroform	Lead	1,2-Dichloroethane	Lead
1,1,1-Trichloroethane	1,2-Dichloroethane	---	1,1,1-Trichloroethane	---
1,1,2-Trichloroethane	1,1,1-Trichloroethane	β-BHC	1,1,2-Trichloroethane	PCBs
Carbon tetrachloride	1,1,2-Trichloroethane	t-Nonachlor	Carbon tetrachloride	
Vinylidene chloride	Carbon tetrachloride	Oxychlordane	Vinylidene chloride	
Trichloroethylene	Vinylidene chloride	Heptachlor epoxide	Trichloroethylene	
Tetrachloroethylene	Trichloroethylene	PCBs	Tetrachloroethylene	
Bromodichloromethane	Tetrachloroethylene	---	Bromodichloromethane	
Chlorobenzene	Bromodichloromethane	Benzo(a)pyrene	Chlorobenzene	
	Chlorobenzene	Fluoranthene	---	
	---	Benzo(k)fluoranthene	Arsenic	
	Arsenic	Pyrene	Lead	
	Cadmium	Chrysene	Cadmium	
	Lead	Benzo(a)anthrene	---	
	---		β-BHC	
	β-BHC		Lindane	
	Lindane		Heptachlor epoxide	
	Heptachlor expoxide		t-Nonachlor	
	t-Nonachlor		Oxychlordane	
	Oxychlordane		HCB	
	HCB		DDT/DDD/DDE	
	DDT/DDD/DDE		PCBs	
	PCBs			

	Benzo(a)pyrene			
	Fluoranthene			
	Benzo(k)fluoranthene			
	Pyrene			
	Chysene			
	Benzo(a)anthrene			

Table 4. SAMPLING FRAMEWORK FOR PHASE I PARTICIPANTS (NEW JERSEY SUBJECTS)

Day	Participant[a] group	Matrix sampled								
		Air/Vap.[b]	Air/Part.[c]	DW[d]	Bev.[e] & Foodstuffs	H. Dust[f]	Blood[g]	Breath[h]	Urine[i]	Hair[j]
1	I	x		x						
2	I	x		x			x	x	x	
3	I	x	x	x	x	x	x	x	x	x
4	II	x		x						
5	II	x		x			x	x	x	
6	II	x	x	x	x	x	x	x	x	x
7	III	x		x						
8	III	x		x			x	x	x	
9	III	x	x	x	x	x	x	x	x	x

[a]Each group consists of three persons.

[b]Samples taken 7:30 AM - 5:30 PM, 5:30-10:30 PM, 10:30-7:30 AM

[c]Samples taken 5:30 PM - 7:30 AM or 7:30 AM - 5:30 PM

[d]Samples taken at home and work.

[e]Samples from supermarket.

[f]Samples from home.

[g,h]Samples taken between 5 and 7:00 PM.

[i]Sample (O.N.) taken AM.

[j]Sample taken from males during haircut.

Abbreviations:

Vap. = vapors
Part. = particulate
DW = drinking water
Bev. = beverages
H. dust = household dust

Results

Volatile Organics. Ten of the 12 target compounds were present in both air and breath samples (Figs. 4-6). Only chloroform and bromodichloromethane were present in drinking water in significant amounts. The food measurements using FDA's headspace methodology indicated that the 40 or so solid food samples tested were relatively unimportant as a source for the target compounds, with the possible exception of trichloroethylene, which was occasionally found at high levels (> 1000 ng/g) in margarine samples. Beverages, particularly cola soft drinks, were significant contributors of chloroform and bromodichloromethane. Blood and urine analytical protocols were inadequate to detect normal (nonoccupational) levels of VOC's.

Metals. Lead was detected in 17 of 18 personal air samples. The median concentration was 0.53 ug/m^3. House-dust samples contained all three metals, but lead was most prevalent, with a median concentration of 440 ug/g. Water samples contained lead at a median value of 5.1 ug/L. Assuming 10 m^3 of air intake per day and 1 L/day of drinking water, median intakes of lead through air and water were nearly identical at 5 ug/day each. However, the food intakes, as measured by a concurrent FDA Market Basket Survey in Elizabeth, were estimated as 35 ug/day. Thus the mean daily food intake so outweighed the air and water intake that individual food samples would have to be collected in order to relate body burden to total daily intake. Since protocols for collecting individual food items from peoples' homes do not exist, metals were removed from consideration for the large-scale survey of exposures planned in Phase II of the TEAM Study.

Lead and cadmium in blood were detected in all 33 samples at median values of 23.5 and 0.75 ug/deciliter, respectively. Interferences invalidated the lead and cadmium analyses of urine samples, but arsenic was detected in 34 of 35 samples at a median value of 9.7 ug/L.

Pesticides and PCB's. The collection and analysis methods for these semivolatile organics in air proved too insensitive to detect normal levels. Toward the end of the study, however, an improved analysis technique was developed. An improved monitor collecting larger volumes of air (6L/min for 24 hours) was also developed and tested in a later field study, together with the improved analytical method. The new combination was found to collect sufficient amounts of indoor airborne pesticides to provide useful measurements. No pesticides were found in water, although the analytical technique could detect concentrations below 1 ug/L. No pesticides were detected in dust. The two analytical procedures tested for blood samples both proved capable of detecting pesticide levels ranging from 0.1 to 0.5 ug/L for most of the target pesticides, agreeing generally with the New Jersey values obtained in US National Center for Health Statistics (NCHS) Health and Nutrition Examination Survey (HANES).

PAH's. The sampling and analytical methods proved inadequate to detect any of these compounds at indoor concentrations. Similar difficulties were encountered in the analyses of PAH's in drinking water, dust, blood, and urine.

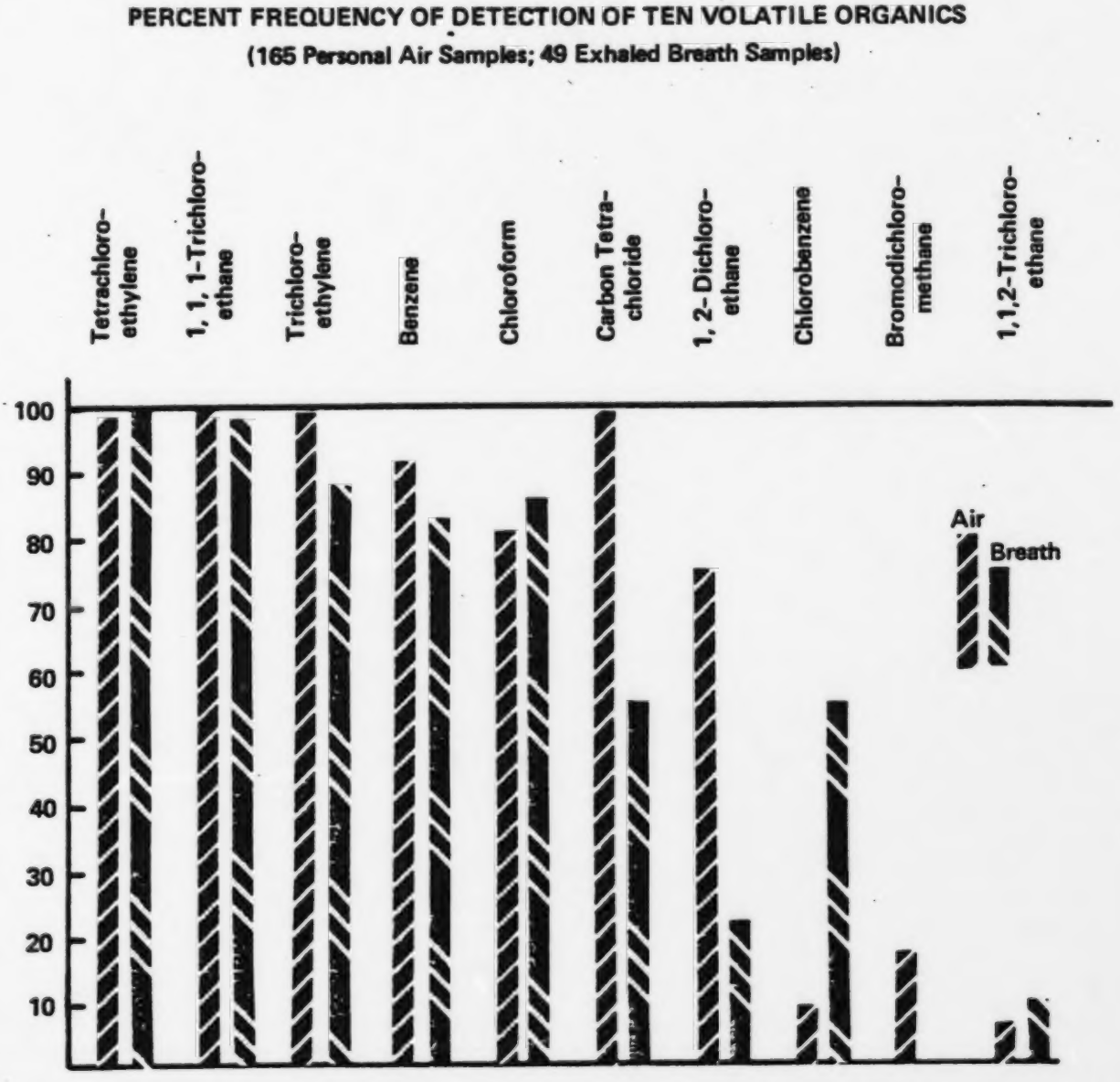

Fig. 4. Ten of 12 target chemicals were found in air or breath samples.

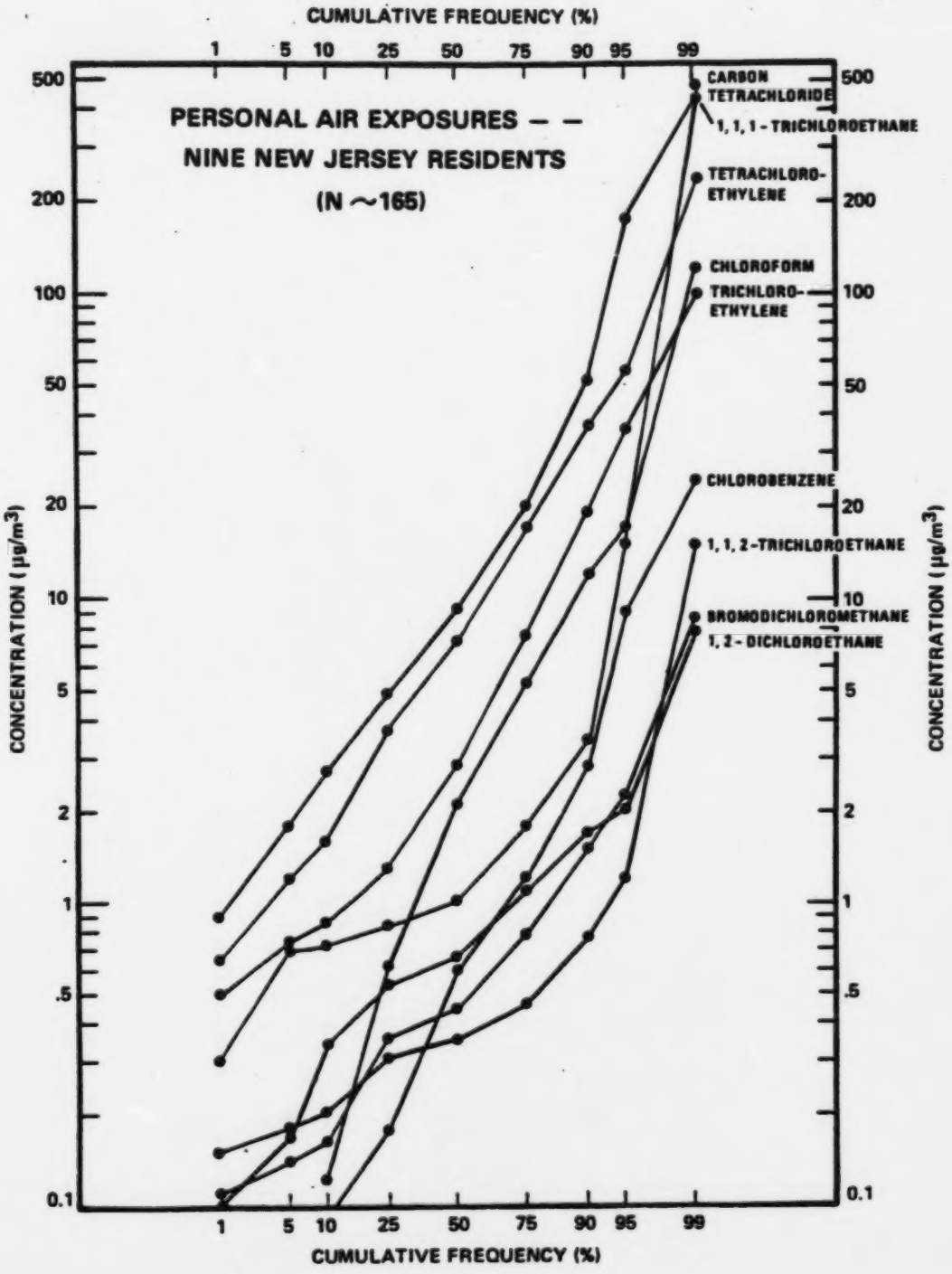

Fig. 5. Distributions of personal air concentrations of nine chemicals displayed departures from log-normality.

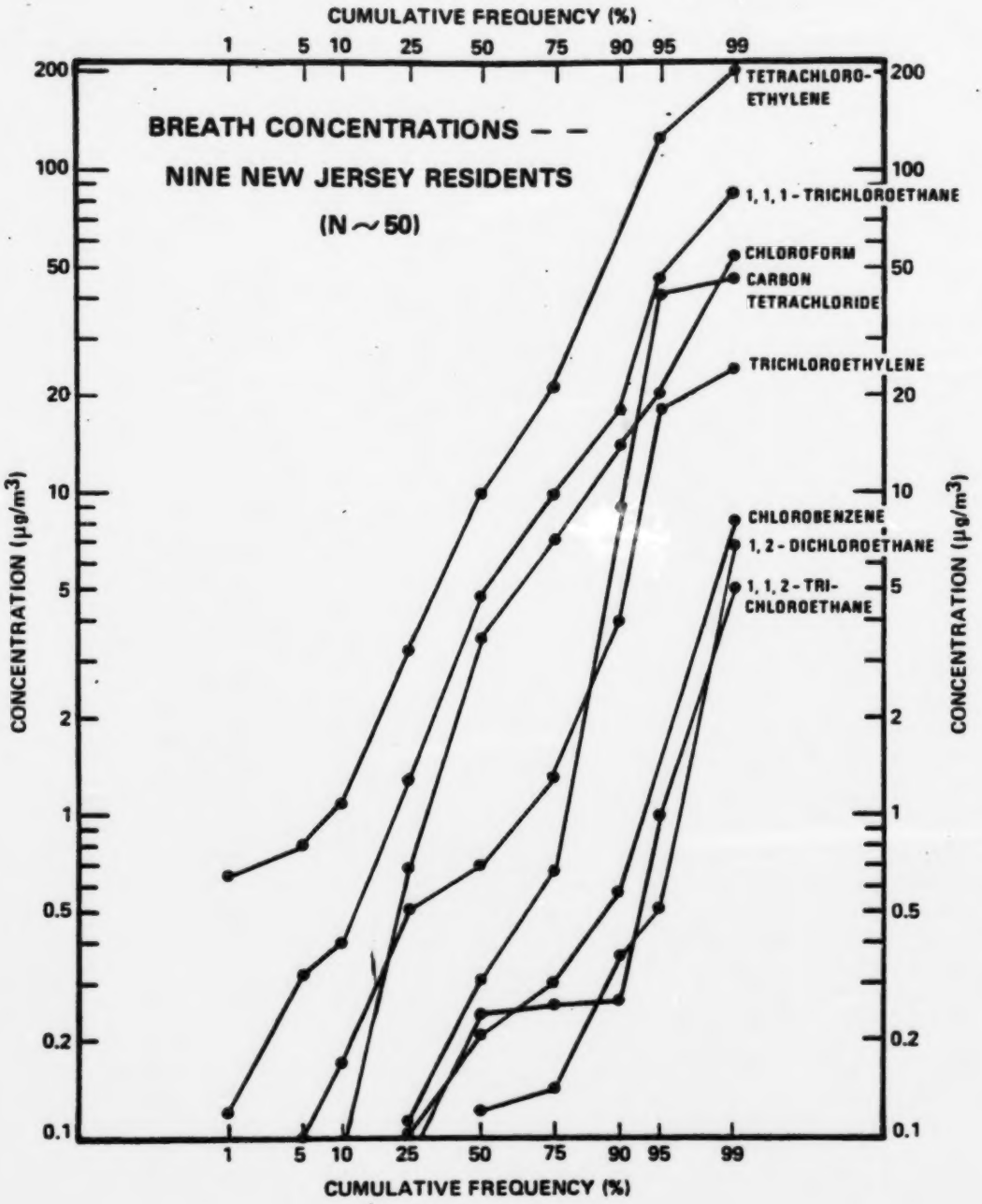

Fig. 6. Breath concentrations also displayed departures from log-normality.

Evaluation of Pilot Study Protocols

Of the four groups of compounds, only the volatile organics were suitable for a large-scale survey of a statistically meaningful sample of the general population. Only air, water, and breath need be sampled, since the exposure through food and beverages is likely to be negligible. (Chloroform is an exception, since significant amounts may be ingested in soft drinks).

For the metals, protocols are needed for sampling and analyzing individual meals prepared at the subject's home before a larger survey could be carried out. Such protocol development was begun in a special study carried out in North Dakota in October of 1982. Initial results indicate that interference problems continue to be encountered in the analysis of metals in solid matrices. If these problems can be resolved, metals could become the second group of pollutants for which the total exposure concept could be field tested.

For the pesticides and PCB's, encouraging progress was made in developing a personal sampler and an analytical method capable of detecting airborne pesticides at indoor concentrations. The method was tested in Greensboro, N.C. on 20 randomly-chosen respondents and pesticide exposure was documented for all 20. However, uncertainties regarding the collection and analysis of individual food items make it currently unprofitable to attempt a total exposure study.

For the PAH's, no personal air sampler exists. Sampling and analytical protocols for measuring personal exposure to these compounds in other environmental and biological media are also unsatisfactory.

Exposure - Body Burden Relationships

A simple equilibrium model relating breathing-zone concentrations A directly to exhaled breath concentrations B ($B = f A$, where f = fraction of chemical exhaled at equilibrium) gave significant ($p < .01$) fits to the data from approximately 60 breath measurements for three chemicals: tetrachloroethylene ($r = .49$); trichloroethylene ($r = .43$), and 1,1,1-trichloroethane ($r = .56$).

Regression parameters were

Tetrachloroethylene: $\log B = 0.87 \pm 0.28 + (0.57 \pm 0.11) \log A$
1,1,1-Trichloroethane: $\log B = -0.78 \pm 0.32 + (0.60 \pm 0.10) \log A$
Trichloroethylene: $\log B = -0.70 \pm 0.25 + (0.35 \pm 0.08) \log A$

The slopes indicated by the regression model are all less than the value of 1 predicted by the equilibrium model. That is, breath values are lower than predicted by the equilibrium model when air exposures are high, and higher than predicted when air exposures are low. This is a result of ignoring the time taken by the body to come to equilibrium. For example, during high exposures, the body is approaching, but has not yet attained, a new (higher) equilibrium concentration; breath samples taken at this time will be lower than predicted by the equilibrium model. Similar reasoning shows that breath samples taken following low exposures will be higher than expected.

A significant correlation was also found between chloroform concentrations in drinking water and in exhaled breath ($r = .48$; $N = 41$; $p<.01$) but not between concentrations in air and breath ($r = -.02$; $N = 60$; $p>.9$), as expected from our estimates that less than 25% of chloroform exposures for these subjects came from air.

Although air was not the main medium of chloroform exposure, an interesting phenomenon was observed in the airborne chloroform exposures of the three North Carolina volunteers: night-time concentrations averaged three times greater than day-time levels, a statistically significant difference. Since day-time samples were usually collected at work or in transit, while night-time samples were nearly always taken in the home, it appears that either indoor air in the subject's homes contained more chloroform than a weighted average of outdoor and workplace atmospheres, or night-time values in homes are greater than day-time values, or both. It may be that the chloroform released from hot showers or by boiling water for dinner created higher overnight levels indoors.

The equilibrium model described above suffers not only from the assumption of constant exposures but also from an inability to account for the effect of the residence time τ in the body of any given target chemical. That is, a step change in the ambient concentration will cause an exponential rise or decay toward a new equilibrium (assuming a single body compartment) according to the equation

$$B = B_o\, e^{-t/\tau} + f\, A\, (1-e^{-t/\tau})$$

where B_o is the breath concentration at the beginning of the exposure period. A more complex behavior involving several different residence times τ_i can be modeled assuming multiple compartments. In the latter case, a "lumped" residence time τ_{EFF} could be determined from observation of the time decay or rise of the breath concentration. Since the sampling protocol included two breath samples taken 24 hours apart, with measurements of the three intervening 8-hour exposures, it was possible to create and test a time-dependent model to determine the effective residence time τ_{EFF} of two chemicals: tetrachloroethylene and 1,1,1-trichloroethane:

$$B = B_o e^{-24/\tau_{EFF}} + f \sum \alpha_i A_i$$

where A_i = average personal exposure during the ith exposure period, and the τ_i are weighting functions depending only on t_i and τ_{EFF}.

A computer search for the "best" values of τ_{EFF} and f by minimizing the sum of the squared residuals (using log-transformed data) resulted in the following values:

Tetrachloroethylene (N=30): τ_{EFF} = 31 hours, f = 1.00,
1,1,1-Trichloroethane (N=29): τ_{EFF} = 13 hours, f = 0.13

Thus essentially all the inhaled tetrachloroethylene ultimately exits via the breath, but only 13% of the inhaled 1,1,1-trichloroethane exits unchanged via the breath. The long half-life of tetrachloroethylene (21 hours) indicates that the influence of the initial breath concentration

B∘ on the final breath concentration B cannot be ignored unless the sampling period is longer than 24 hours; however, the shorter halflife of 9 hours for 1,1,1-trichloroethane means a 24-hour sampling period is sufficient to "wash out" the influence of the initial breath concentration. A recent study (Wallace 1983b) measured τ_{EFF} directly for tetrachoroethylene, by placing a subject (previously exposed for one hour inside a dry cleaning shop) in a clean-air chamber and measuring his exhaled breath at intervals for the following 10 hours to detect the decline in tetrachloroethylene concentrations. The initial breath level of 550 ug/m^3 declined to 350 ug/m^3 after 10 hours, and the best-fit exponential had a slope corresponding to a half-life of 21 hours, in agreement with the value of 21 hours calculated independently from the 12 New Jersey and North Carolina subjects.

Phase II: Large-Scale Field Study

A total of 22 volatile organic compounds were selected for study in Phase II of the TEAM Study. The objective of this phase was to measure exposure through air and drinking water, and excretion rates through exhaled breath, of the target compounds for a statistically valid sample of 375 residents of Elizabeth and Bayonne, NJ. These subjects were selected from an initial screening of 5,579 homes, representing 100 geographic districts stratified by distance from major point sources and socioeconomic status. 4,465 respondents (80%) provided data for all residents in each home on the secondary stratification variables of age, sex, race, potential occupational exposure, hobbies, and smoking status.

The 362 ultimate participants each carried out the following activities:

- Filled out the Study Questionnaire, containing a total of 51 questions on occupation, personal habits, health, diet, and residential characteristics
- Wore (or placed in his/her bedroom) a Tenax monitor for 12 hours beginning in the evening
- Wore or carried another Tenax monitor for 12 hours beginning the next morning
- Collected one drinking water sample during each 12-hour period (at home and at work, if applicable)
- Kept a log of activities during the 24-hour period
- Provided a breath sample at the end of the 24-hour period
- Filled out a 24-hour recall questionnaire concerning 11 exposure variables

In addition, one respondent from each of the 97 districts ultimately sampled allowed an identical Tenax monitor to be set up in his/her backyard to take two consecutive 12-hour outdoor samples during the same 24-hour exposure period. We treated these as "fixed-site" stations to compare with personal exposures.

Results

The Study Questionnaires indicate that a rather high 34% were employed in occupations (chemical manufacture, petroleum products, service stations, bus or taxi drivers) with potential exposure to the target chemicals. The percentage of current smokers (45%) was also well above the national average of 33%. The age ranged from 5 to 89 years, with a median of 31, similar to the national median. The percentage of Hispanics (17%) was much higher than the national value of 6% but the percentage of Blacks (12%) was identical to the national figure. Only 13% reported using pesticides, which seems lower than nationwide estimates.

Responses to questions on health indicated that subjects perceived their health as excellent (29%); good (56%); or fair (15%) -- only 2 persons reported poor health. However, 19% reported current respiratory problems, 22% reported taking prescription medicines and 16% were currently under a doctor's care.

Self-described activity levels were reported as heavy (15%), light (63%) and sedentary (23%). Time spent away from home averaged 8.2 $\pm$ 4.1 hours on weekdays and 6.9 $\pm$ 5.2 hours on weekend days.

From the 24-hour recall questionnaire, subjects reported considerable exposure to occupations or products involving the target chemicals. Meat, vegetable, and fruit consumption was remarkably similar to nationwide patterns, but fish and cheese consumption was at least twice the national figure. Coffee, soft drink, and milk consumption was also similar to national figures.

Nearly 2,000 field samples (air, breath, and water) and over 800 quality control samples were collected from the 362 participants and 97 fixed sites (Table 5).

Indoor-Outdoor Comparisons

Overnight air samples were collected from 97 backyard sites in Elizabeth and Bayonne. These samples were matched with overnight personal air samples collected by the resident of the site. Since many residents did not leave their homes after being issued the personal monitor, it is possible to regard most of the personal samples as representative of the indoor atmosphere of the house between 6 PM and 6 AM.

At least 18 of the 22 target compounds were identified in one or more air samples (Table 6). Two of the four missing compounds -- the meta isomers of dichlorobenzene and xylene -- may also have been present, but could not be separated chromatographically from the para isomers. Only bromoform and dibromochloropropane were never found. Toluene was present in all samples, but was also present in most Tenax blanks; therefore no attempt at quantitation was made.

Six of the remaining 17 compounds were present in quantifiable amounts in less than 10% of the samples. These were vinylidene chloride; 1,2-dichloroethane; bromodichloromethane; dibromochloromethane; chlorobenzene; and o-dichlorobenzene.

Table 5. TEAM STUDY, PHASE II, FIRST SEASON: SAMPLES ANALYZED

	Personal Air	Fixed Air	Exhaled Breath	Drinking Water	Total
Field	671	183	356	717	1927
Duplicates	121	32	34	69	256
Blanks	111	37	72	63	283
Spikes	116	39	69	65	289
Total	1019	291	531	914	2755

Table 6. FREQUENCIES OF DETECTION OF 22[a] VOLATILE ORGANICS IN OVERNIGHT AIR SAMPLES COLLECTED BY A PERSONAL MONITOR AND BY AN OUTDOOR MONITOR (N =97)

	Type[b]	NC[c]	<LOD[d]	Trace[e]	Quant[f]
Vinylidene Chloride	P	2	82	11	2
	0	9	83	4	1
Chloroform	P	2	20	20	55
	0	9	45	6	37
1,2,-Dichloroethane	P	2	84	8	3
	0	9	75	8	5
1,1,1-Trichloroethane	P	2	5	13	77
	0	9	3	12	73
Benzene	P	2	6	1	88
	0	10	11	1	75
Carbon Tetrachloride	P	2	15	56	24
	0	10	3	31	53
Trichloroethylene	P	2	13	36	46
	0	10	7	25	55
Bromodichloromethane	P	2	83	8	4
	0	9	87	0	1
Dibromochloromethane	P	2	95	0	0
	0	9	87	1	0
Tetrachloroethylene	P	2	6	8	81
	0	11	4	16	66

Table 6 (con't)

	Type	NC	<LOD	Trace	Quant
Chlorobenzene	P	2	77	8	10
	0	10	74	8	5
Bromoform	P	2	95	0	0
	0	10	87	0	0
Styrene	P	3	5	11	78
	0	12	28	21	36
m,p-Dichlorobenzene[g]	P	2	6	18	71
	0	10	14	33	40
o-Dichlorobenzene	P	2	63	24	8
	0	11	70	14	2
Ethylbenzene	P	3	2	4	88
	0	12	4	6	75
o-Xylene	P	3	2	8	84
	0	12	2	11	72
m,p-Xylene[g]	P	3	1	0	93
	0	11	2	2	82

[a] Toluene was detected in all samples but was not quantified due to high background on Tenax
[b] p= personal sample; 0=outdoor sample
[c] NC= Not Calculated
[d] <LOD = Below Limit of Detection
[e] Trace = Below Quantifiable Limit
[f] Quant = Above Quantifiable Limit
[g] Meta and para isomers combined

Eleven compounds were present in abundance (Table 7). Median values ranged from 1-10 ug/m^3, except for 1,1,1,-trichloroethane (15 ug/m^3), p-xylene (14.5) and benzene (13). Mean values were much larger (up to 50 ug/m^3) indicating a right-skewed distribution. (Skewness values ranged from +2 to +8 for all compounds). Cumulative frequency distributions on lognormal-probability paper (Fig. 7-8) indicated only a rough approximation to a lognormal curve, with both convex and concave shapes represented.

Discussion

Personal exposures were consistently greater than the outdoor concentrations for all of the eleven chemicals with significant detection frequencies. For example, personal mean, median, and maximum values were greater than outdoor mean, median, and maximum values for all 11 chemicals. The ratio by which the personal median exceeded the outdoor median varied from 1.4 (trichloroethylene) to 4.2 (chloroform) (Table 8). An even greater range of ratios is associated with the highest values. Two compounds -- p-dichlorobenzene and 1,1,1-trichloroethane -- reached maximum values 20-50 times higher in personal than in outdoor samples, indicating a likely indoor source.

Correlations between the personal and overnight air samples were quite low for most chemicals (Table 9). This is further evidence for the unreliability of outdoor monitors in estimating personal exposures to these chemicals.

Frequencies of detection were also considerably greater for the personal than the outdoor air samples for two chemicals: chloroform and styrene. Using outdoor measurements only, up to half the population might be estimated to be unexposed to airborne chloroform, but the estimate from personal measurements would be less than 25%. For styrene, corresponding estimates of the unexposed population would be 33% (outdoor) but only 6% (personal).

Thus, employing fixed stations alone to estimate population exposure, when applied to urban population of hundreds of thousands or millions, could result in serious underestimations of exposure.

Supporting evidence for high personal/outdoor ratios of volatile organic compounds has been obtained in another EPA study (Pellizzari 1983) of halocarbons in Baton Rouge, LA and Greensboro, NC. All eight target compounds in Greensboro and six of eight in Baton Rouge had personal/outdoor median ratios exceeding 1.0, with values reaching as high as 7 (chloroform in Greensboro); 8 (1,1,1,-trichloroethane in Baton Rouge); and 16 (p-dichlorobenzene in Greensboro). Frequencies of detection were also significantly greater for personal samples. For example, if exposures in Greensboro had been estimated from ambient measurements, 35,000 of the 100,000 residents would have been adjudged free of exposure to tetrachloroethylene; but an estimate from the personal exposure data would predict that less than 3000 residents are unexposed.

Conclusions

In the study areas, essentially all subjects were exposed to at least six and probably 10 hazardous organic vapors, as measured both by personal and fixed air samples and confirmed by breath samples. Exposures were highly variable, ranging over several orders of magnitude. All exposures were well below occupational standards, but possible long-term effects of chronic low-level exposures to these compounds may present a significant public health hazard.

For the eleven most prevalent compounds, personal air median concentrations ranged from 40% to 320% higher than the outdoor fixed air concentrations, indicating widespread indoor sources. Correlations between personal and outdoor samples were poor, even though the outdoor samples were taken in the subjects' back yards. This combination of higher personal air concentrations and poor correlations with fixed outdoor samples shows that it is scientifically invalid to estimate exposures to these compounds from outdoor measurements. Personal air, indoor air, or breath measurements are far superior to outdoor measurements in estimating exposures.

The use of breath samples to confirm measurements of exposure is supported by this study. The sensitivity of the method was demonstrated by finding most of the target compounds in the breath of most of the subjects. On the other hand, the use of blood and urine samples for detecting exposure is contraindicated, since only one compound (chloroform) could be detected in most cases.

It is evident that further studies of personal exposure to these and similar compounds need to be caried out in other cities to determine the nationwide dimensions of this problem. It is also clear that studies are needed to determine exactly why and how these elevated levels occur.

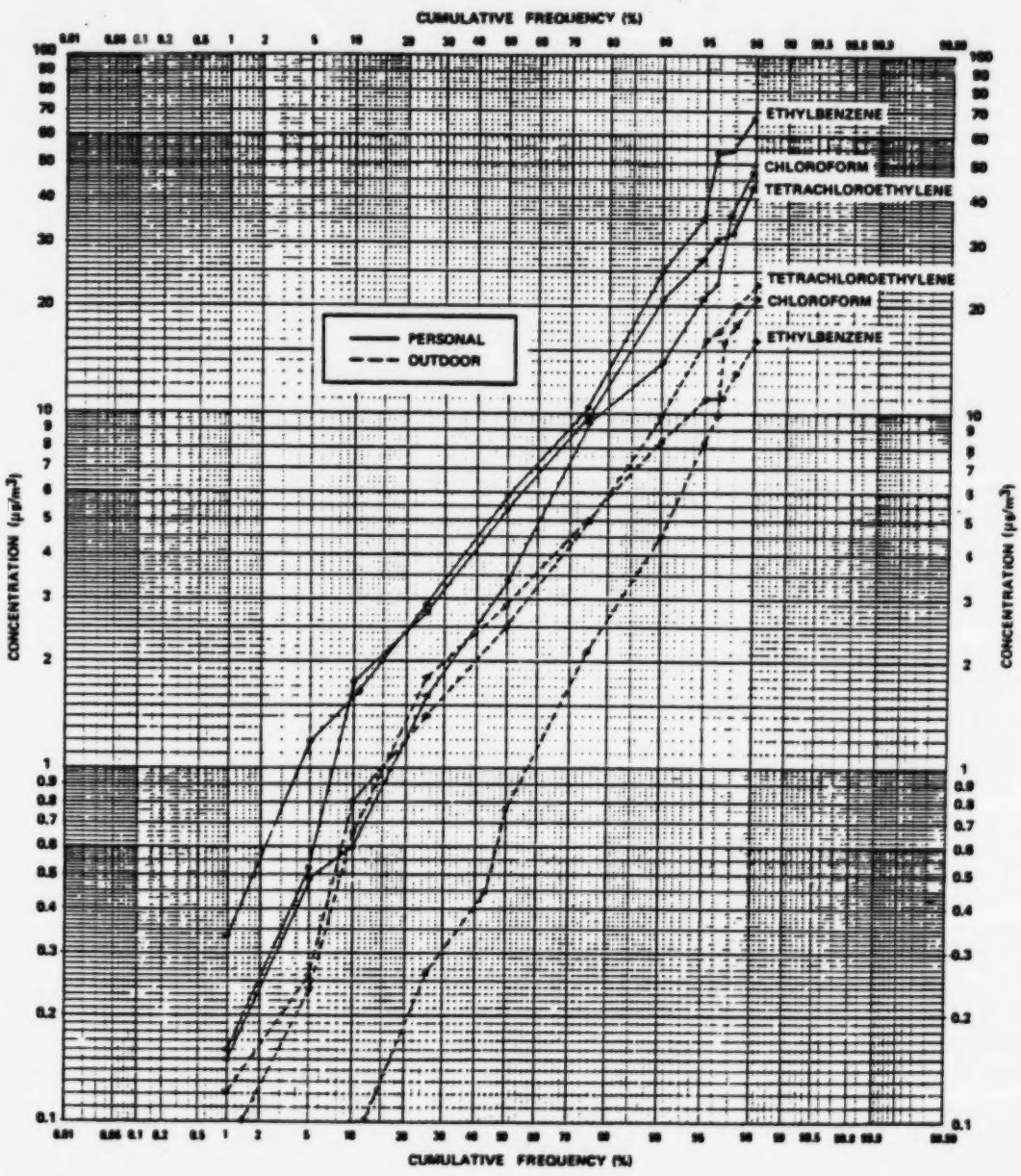

Fig. 7. Personal air concentrations of these chemicals were usually well above the outdoor levels.

Table 7. SUMMARY STATISTICS: MATCHED OVERNIGHT AIR SAMPLES — PERSONAL VS OUTDOOR ELIZABETH AND BAYONNE, NJ

	Type[a]	N[b]	Minimum	Median	Mean(+.S.D.)	Maximum
Benzene	P	95	0.04	13	20 (22)	120
	0	101	0.08	6.6	9.8 (13)	91
Carbon Tetrachloride	P	95	0.5	2.2	2.5 (2.0)	14
	0	101	0.08	1.0	1.4 (1.6)	14
Chloroform	P	95	0.2	3.4	8 (21)	200
	0	102	0.08	0.8	2 (3)	25
m,p-Dichlorobenzene[c]	P	95	0.2	3.4	41 (150)	1200
	0	101	0.1	1.2	1.8 (2.3)	15
Ethylbenzene	P	94	0.3	5.9	13 (34)	320
	0	99	0.08	2.9	3.9 (3.4)	20
Styrene	P	94	0.1	1.9	3.1 (5.9)	54
	0	99	0.1	0.9	1.2 (1.5)	11
Tetrachloroethylene	P	95	0.2	5.4	10 (26)	250
	0	100	0.1	2.5	4.3 (4.9)	27
1,1,1,-Trichloroethane	P	95	0.7	15	50 (125)	880
	0	102	0.09	4.4	6.5 (6.6)	40
Trichlorethylene	P	95	0.3	2.3	3.6 (5.2)	47
	0	101	0.04	1.7	2.7 (2.9)	15
o-Xylene	P	94	0.4	5.2	7.8 (8.6)	49
	0	99	0.4	3.2	4.2 (3.9)	27
m,p-Xylene[c]	P	94	0.4	14.5	21 (24)	120
	0	100	0.3	9.2	11 (9.6)	70

[a] P = personal sample; 0= outdoor (backyard) sample
[b] N includes duplicate samples
[c] meta and para isomers combined

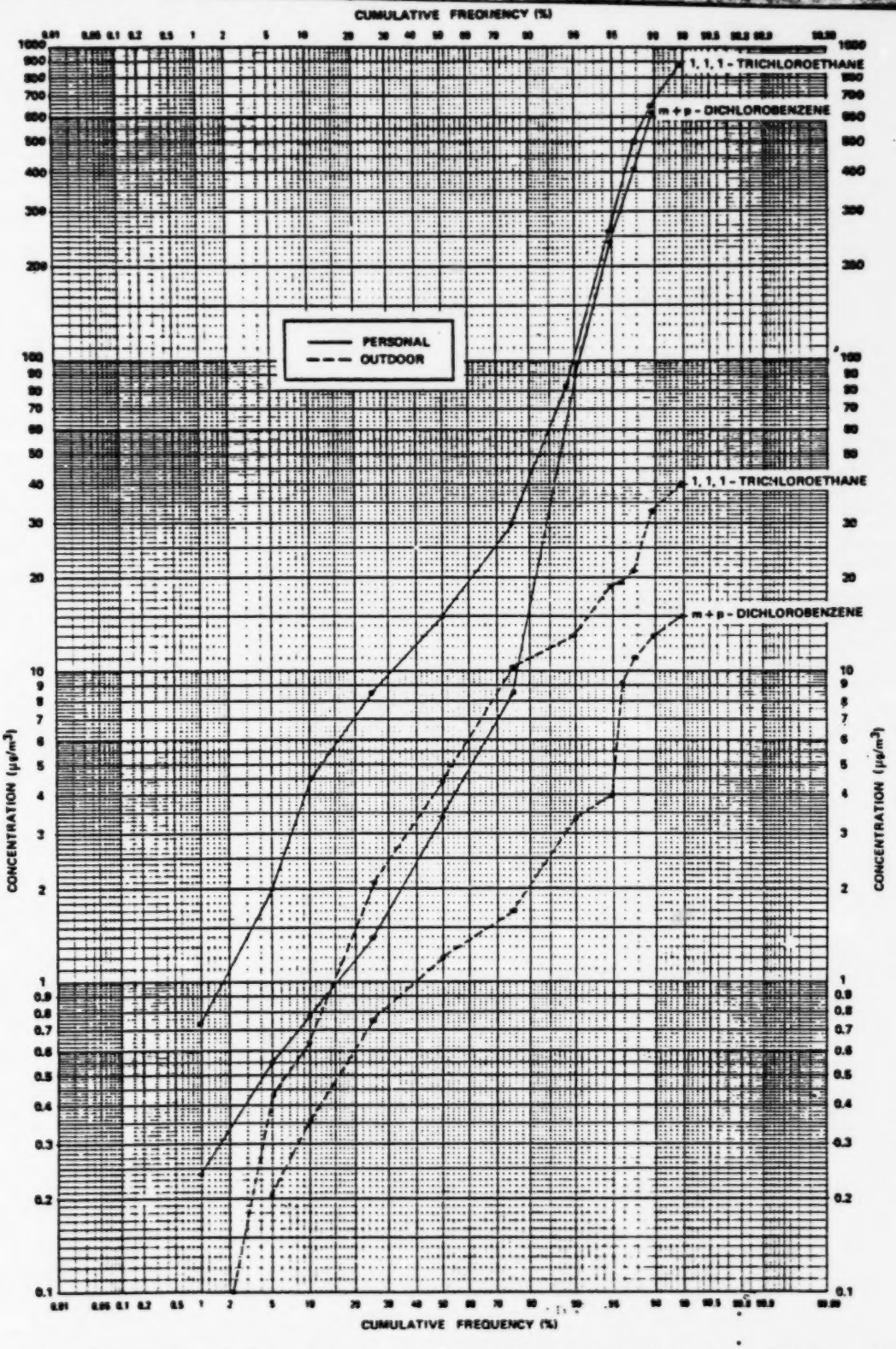

Fig. 8. These two chemicals had the highest personal/outdoor ratios, indicating likely indoor sources.

Table 8. COMPARISON OF OVERNIGHT PERSONAL AIR CONCENTRATIONS WITH OUTDOOR LEVELS OF 11 VOLATILE ORGANIC COMPOUNDS — Elizabeth and Bayonne, NJ

	Mean of Five Highest Values			Median		
	Personal	Outdoor	Ratio	Personal	Outdoor	Ratio
Benzene	91	56	1.6	13	6.6	2.0
Carbon Tetrachloride	8.9	6.5	1.4	2.2	1.0	2.2
Chloroform	65	16	4.0	3.4	0.8	4.2
m,p-Dichloro benzene	570	10.4	55	3.4	1.2	2.8
Ethylbenzene	11	14	0.8	5.9	2.9	2.0
Styrene	20	6.4	3.1	1.9	0.9	2.1
Tetrachloroethylene	77	21	3.7	5.4	2.5	2.2
1,1,1-Trichloroethane	510	26	19	15	4.4	3.4
Trichloroethylene	19	12	1.5	2.3	1.7	1.4
o-Xylene	3.9	16.8	0.2	5.2	3.2	1.6
m,p-Xylene	109	40	2.7	14	9.2	1.6

Table 9. CORRELATIONS BETWEEN OVERNIGHT PERSONAL AIR SAMPLES AND FIXED OUTDOOR (BACKYARD) AIR SAMPLES FOR SELECTED VOLATILE ORGANICS (N 87)

Chemical	Spearman Correlation Coefficient
Benzene	.35*
Carbon tetrachloride	.29*
Chloroform	.17
m,p-Dichlorobenzene	.41*
Ethylbenzene	.22
Styrene	.11
Tetrachloroethylene	.42*
1,1,1-Trichloroethane	.19
Trichloroethylene	.43*
o-Xylene	.24
m,p-Xylene	.26

*Significant at .01 level.

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Disclaimer

Opinions expressed are those of the authors and do not reflect official positions of the U.S. Environmental Protection Agency. Mention of products or brand names does not imply endorsement by the government.

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